



*Handwritten initials and a dollar sign.*

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of: **MINAMIDA, Yukihiro, et al.**

Group Art Unit: 1711

Serial No.: 10/668,964

Examiner: **R.A. Sergent**

Filed: **September 24, 2003**

**P.T.O. Confirmation No.: 1481**

**FOR: SOLVENT-FREE MOISTURE-CURABLE HOT MELT URETHANE RESIN COMPOSITION**

**SUBMISSION OF APPEAL BRIEF**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

August 22, 2006

Sir:

Submitted herewith is an Appeal Brief in the above-identified U.S. patent application.

Also enclosed is a check in the amount of \$500.00 to cover the cost of filing this Appeal Brief. In the event that any additional fees are due with respect to this paper, please charge Deposit Account No. 01-2340.

Respectfully submitted,

**ARMSTRONG, KRATZ, QUINTOS,  
HANSON & BROOKS, LLP**

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**Daniel A. Geselowitz, Ph.D.**

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**PATENT TRADEMARK OFFICE**

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE BOARD OF PATENT APPEALS  
AND INTERFERENCES

APPEAL BRIEF FOR THE APPELLANTS

Ex parte **Yukihiko MINAMIDA, et al.**

**SOLVENT-FREE MOISTURE-CURABLE HOT MELT URETHANE RESIN COMPOSITION**

Serial Number: 10/668,964

Filed: **September 24, 2003**

Group Art Unit: 1711

Examiner: **R. A. Sergeant**

**Daniel A. Geselowitz, Ph.D.**

Registration No. **42,573**

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**FOR: SOLVENT-FREE MOISTURE-CURABLE HOT MELT URETHANE RESIN  
COMPOSITION**

**APPEAL BRIEF PURSUANT TO 37 CFR 41.37**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

August 22, 2006

Sir:

This appeal is filed in response to the rejection of claims in the Office action dated April 3, 2006, under the Notice of Appeal timely filed on June 29, 2006.

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**I. REAL PARTY IN INTEREST**

The real party in interest is Dainippon Ink and Chemicals, Inc., 35-58, Sakashita 3-chome, Itabashi-ku, Tokyo, Japan, as evidenced by the assignment filed and recorded on September 24, 2003.

## **II. RELATED APPEALS AND INTERFERENCES**

Appellant knows of no other prior appeal, pending appeal, interference, or judicial proceedings which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

### **III. STATUS OF CLAIMS**

Claims 1-4 are pending in this application; no claim has been canceled during prosecution.

Claims 1-4 have been rejected and claims 1-4 are under appeal.

#### **IV. STATUS OF AMENDMENTS**

No amendment has been filed subsequent to the last Office action, which was the non-final Office action of April 3, 2006.



## V. SUMMARY OF THE CLAIMED SUBJECT MATTER

Independent claim 1 recites a moisture-curable hot melt urethane resin composition. “Hot melt” is a generic term referring to the property of being solid or viscous at normal temperature, but being melted to form a fluid or liquid when heated, as described on page 7, 5th paragraph, of the specification. The hot melt composition of claim 1 is free from solvents, and when melted to an applicable state and then cooled, exhibits a cohesive power. The hot melt has utility, for example, as a solvent-free adhesive or a coating material.

The composition of claim 1 comprises three components, labeled (A), (B) and (C) (see page 6, lines 14-18).

Component (A) is an isocyanate group-containing hot melt urethane prepolymer. This is prepared by reacting a polyisocyanate with a polyester polyol (see page 9, second and fourth paragraphs), where the polyester polyol comprises a condensate of a polyhydric alcohol and a polybasic acid (page 9, fifth paragraph), and the polybasic acid is a phthalic acid, an isophthalic acid, a terephthalic acid, or a mixture of these (page 10, lines 4-6).

Component (B) is a crosslinking catalyst of general formula (X) in claim 1. This is a morpholine ether-based compound (see page 12, fourth and fifth paragraphs).

Component (C) is at least one sulfur atom-containing organic acid selected from the group consisting of sulfonic acids and sulfinic acids (see page 13, last paragraph).

**VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL**

A. Whether claims 1-4 are unpatentable over Helmeke et al. (U.S. Patent No. 6,844,073) in view of Lee et al. (U.S. Patent No. 4,574,793) or Oertel (*Polyurethane Handbook: Chemistry--Raw Materials--Processing--Applications--Properties*; Hanser Publishers; New York; 1985; pp. 96 and 97), as stated in paragraph no. 3 of the Office action dated April 3, 2006.

## VII. ARGUMENT

**A. Whether claims 1-4 are unpatentable over Helmeke et al. (U.S. Patent No. 6,844,073) in view of Lee et al. (U.S. Patent No. 4,574,793) or Oertel (*Polyurethane Handbook: Chemistry--Raw Materials--Processing--Applications--Properties*; Hanser Publishers; New York; 1985; pp. 96 and 97), as stated in paragraph no. 3 of the Office action dated April 3, 2006.**

1. Basic Summary of the Rejection as stated in paragraphs no. 4 and 5 of the Office action of April 3, 2006

Paragraphs no. 3 and 4 of the Office action of April 3, 2006, are essentially the same in content as in the previous presentation of the rejection in the final Office action of July 1, 2005.

In the rejection as stated in paragraph no. 3 of the Office action of April 3, 2006, the Examiner cites Helmeke et al. for disclosing curable hot melt adhesives wherein the adhesives comprise polyurethane prepolymers derived from diphenylmethane diisocyanate and polyester polyols, and that the polyester is derived from polyols and phthalic acid. The Examiner cites the reference as stating that morpholine ether catalysts are preferred.

In paragraph no. 4 of the Office action, the Examiner states that the Helmeke et al. reference is silent regarding the incorporation of sulfonic acids into the adhesive compositions. The Examiner cites Lee et al. and Oertel et al. as disclosing that sulfonic acids are known inhibitors for isocyanate reactions, especially in the presence of amine catalysts, and states that it would have been obvious to incorporate sulfonic acids into the amine catalyzed polyurethane prepolymer adhesives of Helmeke et al., to extend shelf life, potlife and workability.

2. General summary of further issues in the rejection in the Office action of April 3, 2006.

In response to the first presentation of the rejection at issue, in the final Office action of July 1, 2005, Applicant filed a Response on September 26, 2005, requesting reconsideration of the rejection. In that response, Applicant argued that the present claims yielded effects that are **unexpected** over the prior art, and in support of this argument, referred to data in the Declaration under 37 CFR 1.132 filed on April 13, 2005 ("Declaration I"), which had been submitted in response to a previous rejection. Declaration I had apparently overcome the previous rejection, but in the final Office action dated July 1, 2005, the Examiner had considered Declaration I insufficient to overcome the newly stated rejection.

Applicant then filed a Request for Continued Examination on November 30, 2005, followed by a Supplemental Response on February 27, 2006, with an additional Declaration under 37 CFR 1.132 ("Declaration II"). Applicant amplified the "unexpected results" arguments based on the data in Declaration II. In particular, Applicant argued that the data in Declaration I and Declaration II demonstrate a **synergy** between the three components recited in claim 1, that is: 1) the polyester polyol made using phthalic acid, isophthalic acid or terephthalic acid, 2) the morpholine-based crosslinking agent, and 3) the sulfonic or sulfinic acid; and Applicant argues that this synergy is not expected based on the prior art.

In the Office action dated April 3, 2006, the Examiner addressed Declaration II in paragraph no. 5 of the Office action, stating:

"First, the declaration is not commensurate in scope with the claims. Secondly, the applicants' results are not unexpected. The most relevant comparison stems from a review of Comparative Example 3 and Example 1, where the difference appears to be that Example 1 employs a sulfonate compound ...." (paragraph no. 5 of the Office action)

To clarify the Examiner's position, Applicant conducted a personal interview with the Examiner on June 6, 2006, regarding these two issues raised by the Examiner. No agreement was reached, but the Examiner clarified in the interview why he considered Declaration II to be "not commensurate in scope." Specifically, he explained that the data in the Declaration did not include enough examples of polyisocyanate species, polyol species, and an example of a sulfinic acid, as opposed to a sulfonic acid.

Applicant therefore submits that the main issues in the present appeal are the issues regarding whether the data in Declaration I and Declaration II overcome the rejection. Specifically:

- a) Are the examples in the Declaration an appropriate comparison to the prior art?;
- b) Are the results in the Declaration "unexpected"; and
- c) Are the examples in the Declaration commensurate in scope with the claims?

Applicant here argues that the answer to all three questions is "yes," and that the evidence presented demonstrates that claims 1-4 are not obvious over Helmeke et al., Lee et al. and Oertel, taken separately or in combination.

3. Summary of Appellant's arguments based on Declaration I and Declaration II

The rejection is based on the combination of Helmeke et al., Lee et al. and Oertel. Applicant here first addresses the teachings of these references, and then presents the arguments based on Declarations I and II.

Regarding Helmeke et al.

Helmeke et al. is cited as disclosing curable hot melt polyurethane adhesive comprising polyurethane prepolymers derived from diphenylmethane diisocyanate and polyester polyols, including polyols and phthalic acid. The Examiner also states that morpholine ether catalysts are preferred.

Appellant acknowledges that the general disclosure of Helmeke et al. includes a polyurethane prepolymer that has a polyester polyol made from neopentyl glycol, hexanediol and phthalic anhydride or phthalic acid (abstract).

The Examiner also cites Helmeke et al. at column 4, line 44-56, as stating that morpholine ether catalysts are "preferred." However, Appellant notes in this regard that the catalyst is an optional ingredient in Helmeke et al., since column 4, lines 44-45, state that: "The adhesive composition **may further include** no greater than about 2.0 wt%, preferably from about 0.01wt% to about 2.0 wt% catalyst" (emphasis added). That the morpholine ether catalyst is optional may also be seen from the fact that **all four** of Helmeke's Examples (Examples 1-4 in columns 7 and 8) **do not** have a morpholine ether catalyst. Therefore, although Appellant acknowledges a suggestion for a morpholine ether catalyst, there is **no teaching at all in the reference regarding what results**

**might be expected from the presence of this catalyst**, other than, one would surmise, an increase in reaction rate, since the component is stated to be a "catalyst."

In addition, there clearly is no teaching or suggestion in Helmeke et al. for addition of a sulfonic or a sulfinic acid.

Regarding Lee et al.

Lee et al. discloses polyurethane systems using a tertiary amine catalyst and methane sulfonic acid as a stabilizing agent (column 2, lines 46-50). The methane sulfonic acid is specifically indicated to function as a stabilizing agent to prevent the catalyst from auto-catalyzing alternative reactions to the polyurethane prepolymerization (column 1, lines 43-50). The main observable result of the use of the composition of Lee et al. appears to be excellent shelf life of the prepolymers and the cured products (column 2, lines 35-37). Lee et al. discloses a specific embodiment using bis(2,6-dimethylmorpholino)diethylether as catalyst. This catalyst is indicated to be highly selective in favor of the prepolymer/water reaction, extend shelf life and to provide a quick curing time (column 2, lines 54-61). The combination of bis(2,6-dimethylmorpholino)diethylether and methane sulfonic acid is stated specifically to extend shelf life (column 2, line 60).

Appellant acknowledges Lee's disclosure of the combination of a morpholine ether catalyst and methane sulfonic acid. However, Lee et al. does not disclose or suggest a polyurethane prepolymer made from made from a **polyester** polyol, and therefore there clearly is no suggestion for one which comprises a condensate of a polyhydric alcohol and a **phthalic acid, an isophthalic acid, or a terephthalic acid**. Lee et al. discloses the possible prepolymers at column 6, line 49, to

column 7, line 5, stating: "Of particular importance are prepolymers based on a diphenylmethane diisocyanate, and a polyol mixture of diols and triols; ...," then proceeding to list possible diols and triols. The terms "polyester," "phthalic," "isophthalic" and "terephthalic" do not appear in the reference.

Applicant further notes that there is no suggestion in Lee et al. for use of a sulfinic acid.

Regarding Oertel

Oertel is cited for the general disclosure on pages 96-97 that Brönstedt and Lewis acids are inhibitors of the isocyanate reaction and that p-toluene sulfonic acid can be added at ppm to the isocyanate. Oertel states: "These materials play an important role in **the preparation of prepolymers** based on highly reactive polyols or amines and isocyanates" (emphasis added).

That is, Oertel generally discloses that a Brönstedt or Lewis acid would be used when the polyol or amine was "highly reactive." The p-toluene sulfonic acid is disclosed only as one possible example of a Brönstedt or Lewis acid. There is no disclosure of use of the acid with any particular polyol, and, as noted, the reference indicates that this could be used with amines as well as polyols. Appellant notes that the only suggestion in the reference for use of the Brönstedt or Lewis acid is "in the preparation of the prepolymer," not in further compositions containing the prepolymer.

Applicant's arguments based on evidence in Declaration I and Declaration II

The Examiner's proposed combination of references is therefore based on Helmeke's general polyurethane prepolymer, taken with Helmeke's suggestion for morpholine ether catalysts. The Examiner combines this with Lee's disclosure of a system using a morpholine ether catalyst and methanesulfonic acid, and with Oertel's general disclosure of use of p-toluenesulfonic acid, to add



a sulfonic acid to Helmeke. Appellant notes in this regard that the Examiner's proposed combination of references is **not** based on the modification of any of Helmeke's **Examples** to add a sulfonic acid, since Helmeke's Examples do not have the morpholine ether catalyst.

In arguing against the rejection, Appellant takes the position that **unexpected results** arise from the combination of the **three components recited as (A), (B) and (C)** recited in claim 1. That is, **all three** of these components must be present to see the observed effect, and there is no suggestion in the references for such an effect.

To support this argument, and in supplement to the Examples in the present specification, Appellant has provided experimental evidence in Declaration II to specifically demonstrate the unexpected results.

#### Summary of Declaration II

In Declaration II, data on hot melt urethane resin compositions within the scope of claim 1 (Examples) and Comparative Examples are compared.

Declaration II provides data on two new Examples, listed in the Declaration as Example 1 and Example 2, and a Comparative Example, listed as Comparative Example 3. These data, taken with Example 2 of the specification (listed as Example 3 in Declaration II), and Comparative Examples 1 and 2 of the specification, allow the following comparisons to be made:

Comparison (1) compares Example 1, which has methanesulfonic acid (i.e., component (C)), to Comparative Example 1, which differs only in not having the methanesulfonic acid.

Comparison (2) compares Example 2, having U-CAT2041, the morpholine ether-based crosslinking agent (i.e., component (B)), to Comparative Example 2, which differs only in not having the morpholine ether-based crosslinking agent.

Comparison (3) compares Example 3, having a terephthalate polyester polyol, to Comparative Example (3), which does not have the terephthalate polyester polyol (having only the other polyols).

These Examples and Comparative Examples have been designed to demonstrate in an unbiased manner that the particular effects of the present invention are associated specifically with the presence of all three components, with only the one component omitted in each of the Comparative Examples.

As can be seen in Table 2 and the discussion of the data in Declaration II, the Comparative Examples can **all** be seen to be inferior to **all** of the inventive Examples in the parameter of decline rate of the thermal stability melt viscosity over time, in particular at 36 hours, where two of the Comparative Examples (1 and 2) gelled. In addition, creep resistance on standing was clearly superior for Examples 2 and 3 compared to Comparative Examples 2 and 3.

Appellant therefore submits that the data presented clearly demonstrate an advantageous effect in thermal stability melt viscosity associated only with the presence of all three components (A), (B) and (C).

This effect is clearly **unexpected** over the cited art. Specifically:

Helmeke et al. discloses the use of a polyurethane prepolymer that has a polyester polyol made from a polyol and phthalic acid (corresponding to component (A)), but only suggests a

morpholine ether catalyst (corresponding to component (B)). There is no indication in the reference of **any** effect that would even be associated with the combination of these two components. Since the reference does not disclose use of methane sulfonic acid (or any sulfonic acid), there is no indication of any effect associated with the presence of a sulfonic acid.

Lee et al. discloses the combination of a morpholine ether catalyst and methane sulfonic acid. The advantages associated with that combination are improved "shelf life," that is, the stability of the composition before curing, and a quick curing time. However, there appears to be **no disclosure of any effect on the cured product, and no disclosure of any effect on any parameter related to the thermal stability melt viscosity**. There also clearly is no suggestion in Oertel that any effect would be associated with using a prepolymer based on a polyol and a phthalic, isophthalic or terephthalic acid.

Oertel discloses a general inhibitory effect associated with Brönstedt or Lewis acids, and implies that these are used **in the preparation of the prepolymer** when the polyol or amine is highly reactive. There is no suggestion of any effect on thermal stability melt viscosity of the final product. There is no indication in Oertel that any effect would be associated with a combination with a morpholine ether-based catalyst, or with a prepolymer based on a polyol and a phthalic, isophthalic or terephthalic acid.

Appellant therefore submits that the effects associated with the combination recited in claim 1 are clearly unexpected over the cited art.

4. Arguments addressing the issues raised by the Examiner regarding Declaration II

a) The examples in the Declaration are an appropriate comparison to the prior art

The Examiner states in the Office action dated April 3, 2006, in paragraph no. 6, on page 4, lines 10 and ff., that:

"In other words, applicant's comparative examples must be representative of the closest prior art, specifically, Helmeke et al. With respect to the declaration of February 27, 2006, Comparative Examples (2) and (3) lack components possessed by Helmeke et al. Specifically, Comparative Example (2) **lacks the catalyst** and Comparative Example (3) lacks the phthalic acid based polyester polyol." (emphasis added)

Appellant takes issue with the argument, and submits that the Comparative Examples were fully representative of Helmeke. Most significantly, **Helmeke's actual Examples 1-4 do not contain any morpholine ether catalyst**. The rejection as stated by the Examiner is based solely on the **suggestion** for this catalyst as an optional ingredient, at column 4, lines 44-45. Appellant, in Comparative Example 1, has therefore provided a Comparative Example that is **closer** to the teachings relied on in Helmeke et al. than any of Helmeke's actual examples.

Moreover, Comparative Example (3) lacks the phthalic acid based polyester polyol **specifically** to demonstrate the importance of this component in the presence of the (B) and (C) components.

Appellant therefore submits that the Examiner's assertion the "declaration is insufficient" because it does not compare to the closest prior art, is incorrect.

b) The results in the Declaration are "unexpected"

Appellant has argued above that the Declarations provide evidence of an effect associated only with the presence of all three components (A), (B) and (C), in claim 1. As discussed above, none of the references individually discloses or suggests all three components, and therefore they cannot, even in combination, suggest an effect requiring the presence of all three components. Moreover, as noted above, none of the references discusses the specific parameters discussed in Declaration II: thermal stability melt viscosity over time, and creep resistance of the final product.

In addition, Oertel, one of the references cited for the disclosure of use of a sulfonic acid, discloses the purpose of the sulfonic acid only "in the preparation of prepolymers based on highly reactive polyols or amines and isocyanates," a purpose which has no clear relationship to the effects discussed in the present declaration.

Appellant therefore submits that the results observed for the invention of claim 1 are clearly **unexpected** over the cited art.

c) The examples in the Declaration are commensurate in scope with the claims

This issue, as stated by the Examiner in the Interview Summary of the interview of June 6, 2006, is that the Declarations are not commensurate in scope with the claims "in terms of polyisocyanate species, polyol species, and sulfur atom-containing organic acid species."

Appellant first notes, with regard to the sulfur atom-containing organic acids, that claim 1 recites that component (C) is selected from the group consisting of sulfonic acids and sulfinic acids. The Examiner is stating that the Declarations addressed only the use of sulfonic acids. However, Appellant notes that the Lee et al. reference was cited for the use of methanesulfonic acid, and the

Oertel reference generally discloses "Brönstedt and Lewis acids," with the specific example of p-toluenesulfonic acid. There is no specific suggestion in these references for use of sulfinic acids, and therefore the portion of the scope of claim 1 regarding "sulfinic acids" is **not suggested** by the references. Since there is no suggestion for sulfinic acids, the portion of the claim scope in which component (C) is a sulfinic acid is not obvious over the references, and there is no need for Appellant to address the "sulfinic acid" portion of the claim scope in the Declarations.

The Examiner implies that the Declarations did not provide sufficient examples of polyisocyanate species and polyol species. Claim 1 requires a prepolymer (A) prepared by reacting a polyisocyanate with a polyester polyol which comprises a condensate of a polyhydric alcohol and a polybasic acid, wherein the polybasic acid is selected from the group consisting of a phthalic acid, an isophthalic acid, a terephthalic acid, and a mixture thereof.

However, Example 1 on page 17 of the specification uses 4,4'-diphenylmethane diisocyanate, as compared to Example 2 on pages 17-18 (see Table I on page 20), which uses Isonate 143LJ, and both of these examples show the thermal stability and creep resistance presented above as "unexpected effects" of the present invention. This clearly represents an example of use of a different polyisocyanate with no effect on the "unexpected effects" of the invention.

In Declaration II, all of the inventive and comparative examples have 1,6-hexanediol adipate, 1,6-hexanediol sebacate, and use "Isonate 143LJ," a carbodiimide-modified MDI. All but Comparative Example 3 have neopentyl terephthalate diol. Although it is true that the examples in Declaration II do not use other polyols and polyisocyanates, Declaration II has clearly demonstrated that the observed effects are associated with the combination of components (A), (B) and (C). The

polyols and polyisocyanate used in the examples in the Declaration are quite typical, and Appellant submits that it is completely reasonable to conclude that the "unexpected" effects would be observed with polyols and polyisocyanates other than those used in these examples.

Appellant therefore submits that the Examples in the Declarations, taken with the Examples in the specification, **are** commensurate in scope with the present claims.

Summary

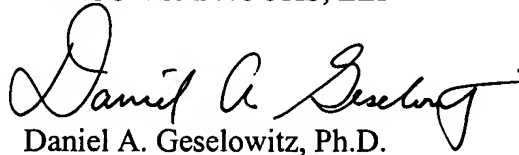
Appellant therefore submits that Appellant has demonstrated unexpected effects associated with the combination of components recited in claim 1, and that the provided examples are commensurate with the claim scope and provide an appropriate comparison to the cited prior art. Claims 1-4 are therefore not obvious over the combination of Helmeke et al. (U.S. Patent No. 6,844,073), Lee et al. (U.S. Patent No. 4,574,793), and Oertel (*Polyurethane Handbook: Chemistry--Raw Materials--Processing--Applications--Properties*; Hanser Publishers; New York; 1985; pp. 96 and 97). Withdrawal of the rejection and allowance of the claims are respectfully requested.

U.S. Patent Application Serial No. 10/668,964  
Appeal Brief

In the event this paper is not timely filed, Appellant hereby petitions for an appropriate extension of time. The fee for any such extension may be charged to our Deposit Account No. 01-2340, along with any other additional fees which may be required with respect to this paper.

Respectfully submitted,

ARMSTRONG, KRATZ, QUINTOS,  
HANSON & BROOKS, LLP



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Appendices: VIII. Claims Appendix  
IX. Evidence Appendix  
X. Related Proceedings Appendix

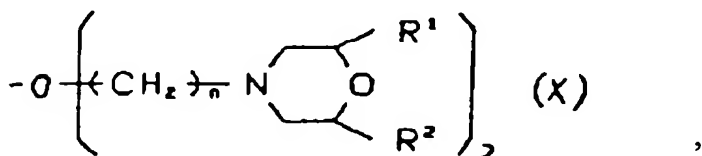


**VIII. CLAIMS APPENDIX**

Claim 1: A solvent-free moisture-curable hot melt urethane resin composition comprising:  
an isocyanate group-containing hot melt urethane prepolymer (A) prepared by reacting a polyisocyanate with a polyester polyol which comprises a condensate of a polyhydric alcohol and a polybasic acid,

wherein the polybasic acid is selected from the group consisting of a phthalic acid, an isophthalic acid, a terephthalic acid, and a mixture thereof;

a crosslinking catalyst (B) of general formula (X):



wherein R<sup>1</sup> and R<sup>2</sup> represent a hydrogen or an alkyl group and n represents a positive integer; and

at least one sulfur atom-containing organic acid (C) selected from the group consisting of sulfonic acids and sulfinic acids.

Claim 2: The solvent-free moisture-curable hot melt urethane resin composition according to claim 1, wherein the isocyanate group-containing hot melt urethane prepolymer (A) is a urethane prepolymer having an isocyanate group, originated from diphenylmethane diisocyanates, at a molecular end of the urethane prepolymer.

Claim 3: The solvent-free moisture-curable hot melt urethane resin composition according to claim 1, wherein the crosslinking catalyst (B) comprises 2,2'-dimorpholino diethyl ether and/or di(2,6-dimethylmorpholinoethyl)ether.

Claim 4: The solvent-free moisture-curable hot melt urethane resin composition according to claim 1, wherein the sulfur atom-containing organic acid (C) comprises methanesulfonic acid and/or ethanesulfonic acid.

\* \* \* \*

**IX. EVIDENCE APPENDIX**

Copies of:

1) the Declaration under 37 CFR 1.132, signed April 12, 2005, and filed with the Amendment dated April 13, 2005 ("Declaration I"; 9 pages), and

2) the Declaration under 37 CFR 1.132, signed February 24, 2006, and filed with the Supplemental Response dated February 27, 2006 ("Declaration II"; 7 pages),  
are attached.



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

Yukihiko MINAMIDA, Masayoshi IMANAKA and Kojiro TANAKA

Application No.: 10/668,964

Group Art Unit: 1711

Filed: September 24, 2003

Examiner: Rabon Sergeant

For: SOLVENT-FREE MOISTURE-CURABLE HOT MELT URETHANE RESIN  
COMPOSITION

DECLARATION UNDER 37 CFR §1.132

COMMISSIONER FOR PATENTS

PO Box 1450

Alexandria

Virginia 22313-1450

Sir:

I, Yukihiko MINAMIDA, hereby declare and state that:

1. I am a citizen of Japan, residing at Dainippon Ink and Chemicals, Inc., 35-58, Sakashita 3-chome, Itabashi-ku, Tokyo, Japan.
2. I am one of the inventors of the subject application, and I am fully familiar with the subject matter thereof as well as the references relied upon by the Examiner in the prosecution of this application.
3. I obtained a Master's degree in Polymer Science and Engineering from the Kyoto Institute of Technology in March 1988.
4. I am currently employed by Dainippon Ink and Chemicals, Inc., and began

working for Dainippon Ink and Chemicals, Inc., on April 1, 1990, in research at the urethane synthesis laboratory. From April 2000, I have engaged in research with functional dispersion technology group.

5. I conducted the following experiments.

### **I. Object**

With regard to the reference (United States Patent No. 4,574,793, United States Patent No. 5,616,625, United States Patent No. 5,550,191) which was cited in the Office Action (dated December 15, 2004) issued on the present application (U.S. Patent Application, No. 10/668,964), tests were conducted to provide supplemental data that the significant effect of the present invention can be obtained only when the polyester polyol having aromatic ring is used in combination with a morpholine ether-based crosslinking catalyst (B), sulfur atom-containing organic acid (C), and that such effect cannot be obtained using the polyol without aromatic ring as disclosed in the reference. It was shown that the polyol without aromatic ring in combination with a morpholine ether-based crosslinking catalyst (B), sulfur atom-containing organic acid (C) disclosed in the reference is not within the scope of claim of the present application.

### **II. Synthesis**

In the following examples, parts are by mass unless otherwise specified.

#### **[Comparative Example 1]**

In four-necked flask, on which a stirrer, a condenser, and a dropping tube were mounted, 100 parts of methyl ethyl ketone, 17.8 parts of butyl methacrylate, 9.4 parts of methyl methacrylate, 0.068 parts of dodecyl mercaptan, and 1.7 parts of azo isobutyro nitrile were heated to 80 °C under stirring. 94.1 parts of butyl methacrylate, 53.6 parts of methyl methacrylate, 0.612 parts of dodecyl mercaptan, and 100 parts of methyl ethyl ketone were added uniformly over a period of 1/2 hour while refluxing by heat of reaction and heating.

275.8 parts of polypropylene glycol having a number-average molecular weight

of 1000, 88.9 parts of polyester polyol having a number-average molecular weight of 3000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, 29.3 parts of polyester polyol having a number-average molecular weight of 1000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid were heated to 100 °C under reduced pressure and then the de-solvent operation was performed until the methyl ethyl ketone was removed.

244 parts of 4, 4—diphenylmethane diisocyanate was added, and the reaction was held at 100 °C. After the 3 hours at 100 °C, 1.2 parts of 2, 2'-dimorpholine diethyl ether (trade name: "U-CAT660M", manufactured by SAN-APRO LTD) was added to obtain a solvent-free reactive hot melt urethane adhesive I having acryl-modified urethane prepolymer as main component. The solvent-free reactive hot melt urethane adhesive I has thermal stability melt viscosity of 9,000mPa·s/125 °C.

#### [Comparative Example 2]

In four-necked flask, on which a stirrer, a condenser, and a dropping tube were mounted, 100 parts of methyl ethyl ketone, 17.8 parts of butyl methacrylate, 9.4 parts of methyl methacrylate, 0.068 parts of dodecyl mercaptan, and 1.7 parts of azo isobutyro nitrile were heated to 80 °C under stirring. 94.1 parts of butyl methacrylate, 53.6 parts of methyl methacrylate, 0.612 parts of dodecyl mercaptan, and 100 parts of methyl ethyl ketone were added uniformly over a period of 1/2 hour while refluxing by heat of reaction and heating.

275.8 parts of polypropylene glycol having a number-average molecular weight of 1000, 88.9 parts of polyester polyol having a number-average molecular weight of 3000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, 29.3 parts of polyester polyol having a number-average molecular weight of 1000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid were heated to 100 °C under reduced pressure and then the de-solvent operation was performed until the methyl ethyl ketone was removed.

250 parts of 4, 4-diphenylmethane diisocyanate was added, and the reaction was held at 100 °C. After the 3 hours at 100 °C, 1.2 parts of U-CAT660M and 0.4 parts of methane sulfonate were added to obtain a solvent-free reactive hot melt

urethane adhesive II having acryl-modified urethane prepolymer as main component. The solvent-free reactive hot melt urethane adhesive II has thermal stability melt viscosity of 7,000mPa·s/125 °C.

[Comparative Example 3]

In four-necked flask, on which a stirrer, a condenser, and a dropping tube were mounted, 100 parts of methyl ethyl ketone, 17.8 parts of butyl methacrylate, 9.4 parts of methyl methacrylate, 0.068 parts of dodecyl mercaptan, and 1.7 parts of azo isobutyronitrile were heated to 80 °C under stirring. 94.1 parts of butyl methacrylate, 53.6 parts of methyl methacrylate, 0.612 parts of dodecyl mercaptan, and 100 parts of methyl ethyl ketone were added uniformly over a period of 1/2 hour while refluxing by heat of reaction and heating.

275.8 parts of polypropylene glycol having a number-average molecular weight of 1000, 88.9 parts of polyester polyol having a number-average molecular weight of 3000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, 29.3 parts of polyester polyol having a number-average molecular weight of 1000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, and 380 parts of an aromatic polyester polyol obtained by using an isophthalic acid and a terephthalic acid (trade name: "Dynacoll 7130", manufactured by SAN-APRO LTD) were heated to 100 °C under reduced pressure and then the de-solvent operation was performed until the methyl ethyl ketone was removed.

300 parts of 4, 4-diphenylmethane diisocyanate was added, and the reaction was held at 100 °C. After the 3 hours at 100 °C, 1.9 parts of U-CAT660M was added to obtain a solvent-free reactive hot melt urethane adhesive III having acryl-modified urethane prepolymer as main component. The solvent-free reactive hot melt urethane adhesive III has thermal stability melt viscosity of 14,000mPa·s/125 °C.

[Example]

In four-necked flask, on which a stirrer, a condenser, and a dropping tube were mounted, 100 parts of methyl ethyl ketone, 17.8 parts of butyl methacrylate, 9.4 parts of methyl methacrylate, 0.068 parts of dodecyl mercaptan, and 1.7 parts of azo isobutyro

nitrile were heated to 80 °C under stirring.

94.1 parts of butyl methacrylate, 53.6 parts of methyl methacrylate, 0.612 parts of dodecyl mercaptan, and 100 parts of methyl ethyl ketone were added uniformly over a period of 1/2 hour while refluxing by heat of reaction and heating. 275.8 parts of polypropylene glycol having a number-average molecular weight of 1000, 88.9 parts of polyester polyol having a number-average molecular weight of 3000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, 29.3 parts of polyester polyol having a number-average molecular weight of 1000 obtained by reacting 1,6-hexane diol, neopentyl glycol, and adipic acid, and 380 parts of Dynacoll 7130 were heated to 100 °C under reduced pressure and then the de-solvent operation was performed until the methyl ethyl ketone was removed.

300 parts of 4, 4-diphenylmethane diisocyanate was added, and the reaction was held at 100 °C. After the 3 hours at 100 °C, 1.9 parts of U-CAT660M and 0.5 parts of methane sulfonate were added to obtain a solvent-free reactive hot melt urethane adhesive IV having acryl-modified urethane prepolymer as main component. The solvent-free reactive hot melt urethane adhesive IV has thermal stability melt viscosity of 12,000mPa·s/125 °C.

#### [Comparative Example 4]

In a 1 liter four-necked flask, 700 parts of long chain aliphatic polyester polyol having a number-average molecular weight of 3500 obtained by reacting 1,2-dodecane dicarboxylic acid with 1,6-hexanediol were heated to 100 °C under reduced pressure and then dehydrated until the water content was reduced to 0.05%.

After cooling to 70 °C, 120 parts of diphenylmethane diisocyanate was added and the mixture was heated to 100 °C and was reacted for 3 hours while being maintained until the isocyanate group content became constant to obtain an aliphatic polyester-urethane prepolymer.

After the completion of the reaction, 1.6 parts of U-CAT 660M and 0.4 parts of methane sulfonic acid were added and stirred uniformly to obtain a solvent-free moisture-curable hot melt urethane resin composition V having an aliphatic polyester-urethane prepolymer as main component. Properties of the resulting



solvent-free moisture-curable hot melt urethane resin composition V has thermal stability melt viscosity of 15,000mPa·s/125 °C.

[Comparative Example 5]

In a 1 liter four-necked flask, 700 parts of polypropylene glycol having a number-average molecular weight of 3000 was heated to 70 °C under reduced pressure and then dehydrated until the water content was reduced to 0.05%.

100 parts of carbodiimide-modified diphenylmethane diisocyanate was added and the mixture was heated to 100 °C and was reacted for 3 hours while being maintained until the isocyanate group content became constant to obtain an aliphatic polyether-urethane prepolymer.

After the completion of the reaction, 1.6 parts of U-CAT 660M and 0.4 parts of methane sulfonic acid were added and stirred uniformly to obtain a solvent-free moisture-curable hot melt urethane resin composition VI having an aliphatic polyether-urethane prepolymer as main component. Properties of the resulting solvent-free moisture-curable hot melt urethane resin composition VI has thermal stability melt viscosity of 1,000mPa·s/125 °C.

### III. Tests

[Method for evaluation of thermal stability]

A solvent-free moisture-curable hot melt urethane resin composition was charged in a metal can having an inner volume of 200 ml and allowed to stand in an oven at 120 °C in a sealed state for 18 hours and 36 hours. Then, it was visually observed whether gelation occurred or not. In the case in which no gelation occurred after standing, the melt viscosity (mPa·s) at 125 °C was measured.

[Method for evaluation of creep resistance at high temperature]

The creep resistance at high temperature was evaluated by the following procedure in place of the evaluation of the moisture-crosslinking reactivity at room temperature.

Using a specimen made by laminating an olefin decorative sheet and a middle

density fiber (MDF) board via a solvent-free moisture-curable hot melt urethane resin composition, the creep resistance at high temperature was evaluated in the following manner.

A solvent-free moisture-curable hot melt urethane resin composition melted heat-melted on a hot plate having a surface temperature of 80°C was applied at a thickness of 40  $\mu\text{m}$  on a 50  $\mu\text{m}$  thick olefin decorative sheet and a MDF board was laminated on the coated surface and, after roll pressing at a linear pressure of 10 kg/cm, the resulting laminate was cut into a width of 25 mm to prepare for a bonded specimen.

A heat-resistant creep test was carried out by allowing specimens, which were allowed to stand for 1, 2, 3, or 4 hours immediately after production at room temperature (23 °C, relative humidity: 65%), to stand in an oven at 60°C for 1 hour while perpendicularly suspending a load (500 g/25 mm) at one end of the olefin decorative sheet in a 90°C angle peeling direction. Then, a creep distance (mm) from the initial suspending location was measured over time. Specimens which had the creep distance of no more than 5 mm were rated “good”.

Since the initial creep properties at 60 °C are improved due to proceeding of the crosslinking reaction at room temperature (23 °C, relative humidity: 65%), the initial creep properties were evaluated in place of the moisture-crosslinking reactivity at room temperature.

#### **IV. Test Results**

The test results are shown in the following Table.1.

Table.1

	Comparative Example 1	Comparative Example 2	Comparative Example 3	Example 1	Comparative Example 4	Comparative Example 5
Thermal stability						
Melt viscosity (mPa·s, 125°C) initial	9,000	7,000	14,000	12,000	15,000	1,000
after 18 hours	9,400	8,100	22,000	14,000	15,000	1,000
after 36 hours	10,300	9,600	Gelled	22,000	19,000	2,000
Creep resistance at 60°C						
Immediately after production of specimen	Fell off	Fell off	Fell off	Fell off	Fell off	Fell off
1 hour after standing at room temperature	Fell off	Fell off	Fell off	Fell off	Fell off	Fell off
2 hours after standing at room temperature	Fell off	Fell off	Fell off	Fell off	Fell off	Fell off
3 hours after standing at room temperature	Fell off	Fell off	26mm	35mm	Fell off	Fell off
4 hours after standing at room temperature	Fell off	Fell off	Good	Good	Fell off	Fell off
6 hours after standing at room temperature	Fell off	Fell off	Good	Good	Fell off	Fell off
8 hours after standing at room temperature	Fell off	Fell off	Good	Good	Fell off	Fell off
12 hours after standing at room temperature	Good	Fell off	Good	Good	Fell off	Fell off

\*Regarding property of creep resistance at high temperature of Comparative Example 4 and 5, low adhesive strength cause "Fell off".

## V. Conclusion

As indicated above, the solvent-free moisture-curable hot melt urethane resin composition, in which the polyester polyol having aromatic ring is used in combination with a morpholine ether-based crosslinking catalyst (B), sulfur atom-containing organic acid (C), has significant effects as claimed in claim 1 of the present application.

We believe, in addition to the significant structural difference between the present invention and the reference, that it is demonstrated the urethane prepolymer disclosed in the reference is not within the scope of claim 1 of the present application.

6. I understand fully the content of this declaration.

7. The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

8. Further declarant saith not.

Yukihiko Minamida

Yukihiko MINAMIDA

2005. 4. 12

Date



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

Yukihiko MINAMIDA, Masayoshi IMANAKA and Kojiro TANAKA

Application No.: 10/668,964

Group Art Unit: 1711

Filed: September 24, 2003

Examiner: Rabon Sergent

For: SOLVENT-FREE MOISTURE-CURABLE HOT MELT URETHANE RESIN  
COMPOSITION

DECLARATION UNDER 37 CFR §1.132

COMMISSIONER FOR PATENTS

PO Box 1450

Alexandria

Virginia 22313-1450

Sir:

I, Yukihiko MINAMIDA, hereby declare and state that:

1. I am a citizen of Japan, residing at Dainippon Ink and Chemicals, Inc., 35-58, Sakashita 3-chome, Itabashi-ku, Tokyo, Japan.
2. I am one of the inventors of the subject application, and I am fully familiar with the subject matter thereof as well as the references relied upon by the Examiner in the prosecution of this application.
3. I obtained a Master's degree in Polymer Science and Engineering from the Kyoto Institute of Technology in March 1988.
4. I am currently employed by Dainippon Ink and Chemicals, Inc., and began working for Dainippon Ink and Chemicals, Inc., on April 1, 1990, in research at the

urethane synthesis laboratory. Since April 2000, I have been engaged in research with the functional dispersion technology group.

5. I conducted the following experiments.

### **I. Object**

Tests were conducted to provide supplemental data showing that the significant effect of the present invention can be obtained only when the polyester polyol having an aromatic ring is used in combination with a morpholine ether-based crosslinking catalyst (B), sulfur atom-containing organic acid (C), and that such an effect cannot be obtained if any one of the polyester polyol having an aromatic ring, a morpholine ether-based crosslinking catalyst (B), or sulfur atom-containing organic acid (C) is lacking. It was shown that the art which does not disclose any one of the polyester polyol having an aromatic ring, a morpholine ether-based crosslinking catalyst (B), or sulfur atom-containing organic acid (C) does not suggest or teach such an effect.

### **II. Synthesis**

In the following examples, parts are by mass unless otherwise specified. According to the formulation shown in the following Table 1, a urethane prepolymer was synthesized by the same operation as in Example 1 of the specification. After the completion of the reaction, additives shown in the following Table 1 were added in the amount described in the same table.

Table 1

Composition of prepolymer (Parts by mass)	Comparison (1)		Comparison (2)		Comparison (3)	
	Comparative Example 1	Example 1	Comparative Example 2	Example 2	Comparative Example 3	Example 3
1,6-hexanediol adipate diol; equivalent weight of OH group=1500	500	500	500	500	500	500
Neopentyl glycol terephthalate diol; equivalent weight of OH group=500	250	250	250	250	-	250
1,6-hexanediol sebacate; equivalent weight of OH group=1750	250	250	250	250	250	250
Isonate 143LJ (carbodiimide-modified MDI, manufactured by Mitsubishi Chemical Corporation)	240	240	240	240	100	240
Methanesulfonic acid	-	0.625	0.5	0.55	1.2	1.2
U-CAT2041	5.0	5.0	-	2.2	9.9	9.9
Melt viscosity (mPa·s, 125°C)	14000	13000	15000	11000	8000	10000
Isocyanate group content (% by weight)	2.1	2.2	2.1	2.2	2.3	2.2

### III. Tests

An evaluation of creep resistance at high temperature and an evaluation of thermal stability are conducted by the same method described in the specification.

#### IV. Test Results

The test results are shown in the following Table.2.

Table.2

	Comparison (1)		Comparison (2)		Comparison (3)	
	Comparative Example 1	Example 1	Comparative Example 2	Example 2	Comparative Example 3	Example 3
Thermal stability Melt viscosity (mPa·s,125°C) initial	12000	11000	13000	11000	8000	12000
after 18 hours	22000	17000	29000	16000	9000	22000
after 36 hours	Gelled	23000	Gelled	20000	12000	35000
Creep resistance at 60°C Immediately after production of specimen	Fell off	Fell off	Fell off	Fell off	Fell off	Fell off
1 hour after standing at room temperature	20 mm	Fell off	Fell off	Fell off	Fell off	9 mm
2 hours after standing at room temperature	Good	Good	Fell off	9 mm	Fell off	Good
3 hours after standing at room temperature	Good	Good	Fell off	Good	Fell off	Good
4 hours after standing at room temperature	Good	Good	Fell off	Good	Fell off	Good

Regarding comparison (1):

Comparison (1) is a comparison between a composition having a urethane prepolymer with an aromatic ring, a morpholine ether-based crosslinking catalyst, and sulfonic acid (Example 1) and a composition having a urethane prepolymer with an aromatic ring, a morpholine ether-based crosslinking catalyst (Comparative Example 1).

As a result, in comparison with the thermal stability melt viscosity of the



composition of Comparative Example 1, that of the composition of Example 1 declined from 12000 mPa·s to 11000 mPa·s (decline rate 8.3 %) initially, from 22000 mPa·s to 17000 mPa·s (decline rate 23 %) after 18 hours, and from the gelled state, in which the viscosity can not be measured, to 23000 mPa·s after 36 hours. Therefore, as time passes, the decline rate of the thermal stability melt viscosity of the present invention is great. Also, especially after 36 hours, it is useful that the thermal stability melt viscosity remarkably declines to a state in which the viscosity can be measured.

Also, from the result of creep resistance of the composition of Example 1, it is shown that the composition of Example 1 has sufficient function as an adhesive.

Therefore, due to the existence of sulfonic acid, the composition of the present invention has a sufficient effect of thermal stability melt viscosity and creep resistance

Regarding comparison (2):

Comparison (2) is a comparison between a composition having a urethane prepolymer with an aromatic ring, a morpholine ether-based crosslinking catalyst, and sulfonic acid (Example 2) and a composition having a urethane prepolymer with an aromatic ring and sulfonic acid (Comparative Example 2).

As a result, in comparison with the thermal stability melt viscosity of the composition of Comparative Example 2, that of the composition of Example 2 declined from 13000 mPa·s to 11000 mPa·s (decline rate 15 %) initially, from 29000 mPa·s to 16000 mPa·s (decline rate 45 %) after 18 hours, and from the gelled state, in which the viscosity can not be measured, to 20000 mPa·s after 36 hours. Therefore, as time passes, the decline rate of the thermal stability melt viscosity of the present invention is great. Also, especially after 36 hours, it is useful the that thermal stability melt viscosity remarkably declines to a state in which viscosity can be measured.

Also, from the result of creep resistance of the composition of Example 2, it is shown that the composition of Example 2 has sufficient function as an adhesive. In contrast, it is shown that the composition of Comparative Example 2 lacks sufficient function as an adhesive

Therefore, due to existence of a morpholine ether-based crosslinking catalyst, the composition of the present invention has a sufficient effect of thermal stability melt

viscosity and creep resistance

Regarding comparison (3):

Comparison (3) is a comparison between a composition having a urethane prepolymer with an aromatic ring, a morpholine ether-based crosslinking catalyst, and sulfonic acid (Example 3) and a composition having a urethane prepolymer without an aromatic ring, a morpholine ether-based crosslinking catalyst, and sulfonic acid (Comparative Example 3).

As a result, the composition of Example 3 has sufficient thermal stability melt viscosity which is a little higher than those of Examples 1 and 2. Also, from the result of creep resistance of the composition of Example 3, it is shown that the composition of Example 3 has sufficient function as an adhesive.

In contrast, the thermal stability melt viscosity of the composition of Comparative Example 3 is lower than that of Example 1. However, from the result of creep resistance of the composition of Example 3, it is shown that the composition of Comparative Example 3 lacks sufficient function as an adhesive.

Therefore, due to the existence of an aromatic ring in a urethane prepolymer, the composition of the present invention has a sufficient effect of thermal stability melt viscosity and creep resistance

## **V. Conclusion**

As indicated above, the solvent-free moisture-curable hot melt urethane resin composition, in which the polyester polyol having aromatic ring is used in combination with a morpholine ether-based crosslinking catalyst (B), sulfur atom-containing organic acid (C), has significant effects as claimed in claim 1 of the present application.

We believe that the art which does not disclose any one of the polyester polyol having aromatic ring, a morpholine ether-based crosslinking catalyst (B), or sulfur atom-containing organic acid (C) does not suggest or teach such an effect.

6. I understand fully the content of this declaration.

7. The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed

to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

8. Further declarant saith not.

Yukihiko Minamida  
Yukihiko MINAMIDA

2006. 2. 24  
Date

**X. RELATED PROCEEDINGS APPENDIX**

As stated in Section (II), there is no related proceeding.